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# NEW HEAVY SCINTILLATING MATERIALS FOR PRECISE HETEROGENEOUS EM-CALORIMETERS

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#### Abstract

Britvich G.I., Britvich I.G., Vasil'chenko V.G. et al. New Heavy Scintillating Materials for Precise Heterogeneous EM-Calorimeters: IHEP Preprint 98-71. – Protvino, 1998. – p. 16, figs. 9, tables 2, refs.: 31.

The investigation results of some optical and scintillation characteristics of new scintillating media based on heavy composite materials and an inorganic crystal CsI : Br, intended for the creation of precise heterogeneous EM-calorimeters with the energy resolution  $\sigma/E \cong 4 - 5\%/\sqrt{E}$ , are presented. The possibility to use cheap heavy scintillating plates based on optical ceramics as active media in heterogeneous EM-calorimeters is considered.

#### Аннотация

Бритвич Г.И., Бритвич И.Г., Васильченко В.Г. и др. Новые тяжелые сцинтиллирующие среды для прецизионных гетерогенных ЕМ-калориметров: Препринт ИФВЭ 98-71. – Протвино, 1998. – 16 с., 9 рис., 2 табл., библиогр.: 31.

В работе представлены результаты исследований оптических и сцинтилляционных характеристик новых тяжелых сцинтиллирующих сред на основе композиционных материалов и неорганического сцинтиллятора CsI : Br, предназначенных для создания гетерогенных ЭМ-калориметров с высоким энергетическим разрешением  $\sigma/E \simeq 4 - 5\%/\sqrt{E}$ . Обсуждаются возможности использования дешевых оптических керамик в качестве активной среды в гетерогенных ЭМ-калориметрах.

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#### 1. Introduction

In recent years, the necessity emerged for the development of electromagnetic (EM) calorimeters which are capable to work under high counting rates to  $10^7 \text{ s}^{-1}$  and high secondary particle multiplicity conditions, display high levels of radiation resistance  $\geq 100 \text{ kGy/yr}$  [1] and energy resolution  $\sigma/E \cong 4-5\%/\sqrt{E}$  [2,3], etc. Nowadays, appreciable attention is given to an opportunity to develop some cheap heterogeneous EM-calorimeters, so-called sampling EM-calorimeters, with a fiber-optic readout system. Such calorimeters can improve the present level of their energy resolution to a level which is characteristic to homogeneous ones. So, it was proposed [4] to use heavy scintillators for the improvement of the detection efficiency of soft  $\gamma$ -quanta in sampling EM-calorimeters, and consequently for the improvement of the energy resolution of modern EM-calorimeters [5,6,7]. The essential features required of heavy scintillators to improve the energy resolution of sampling EM-calorimeters, of "Bayan" [6] and "Shashlyk" [5] types, to a level  $\sigma/E \cong 4 - 5\%/\sqrt{E}$  are formulated in [8]. Thus, our characteristics simulation of some sampling EM-calorimeters [5,6] containing heavy materials as active media with the help of the GEANT program [9] has shown that in the process of transition from ordinary plastic scintillators, with the radiation length  $X_o \cong 42.4$  cm, to heavy materials, with  $X_o \cong 15-20$  cm, the energy resolution might be improved 1.3-1.5 times and would reach a level of  $\sigma/E \cong 4-5\%/\sqrt{E}$ . It is supposed that the light output from new materials under EM-showers excitation should be enough for the fluctuation of the number of photoelectrons registered not to influence the energy resolution of EM-calorimeters.

In the general case, the energy resolution dependence  $\sigma/E$  for sampling EM-calorimeters from the incoming particles energy E can be precisely enough expressed by the formula

$$\sigma/E = a/\sqrt{E \oplus b/E \oplus c},\tag{1}$$

where  $\oplus$  represents quadratic addition, a is the stochastic term determined by the fluctuation of the deposited energy; b is is the so-called "noise" term determined by the used photodetector noise; and c is the constant term determined by light collection nonuniformity from scintillating plates, light output losses in scintillating plates and wavelength shifting (WLS) fibers, the instability of photodetectors, etc. It was experimentally shown that the constant term for the Bayan-type sampling EM-calorimeters could be easily made with c < 0.5% [5]. Note that bvalue gives an appreciable contribution to formula (1) only when silicon photodiodes are used as photodetectors. The energy resolution for the well-designed sampling EM-calorimeters [5-7] is basically determined by the stochastic term a. Thus, the contribution of the sampling fluctuations to expression (1) can be approximately presented in the following way [10]:

$$\sigma/E \propto \sqrt{t_{sc}/f_s},\tag{2}$$

where  $f_s$  is the sampling fraction and  $t_{sc}$  is the thickness of scintillating plates expressed in radiation lengths. Note that most of the well-designed sampling EM-calorimeters with  $\sigma/E < 10\%/\sqrt{E}$  had sampling fractions  $f_s > 0.2$  [10]. Expression (2) also displays two opportunities for the improvement  $\sigma/E$  in sampling EM-calorimeters; the former is connected with the increase  $f_s$  and the latter is connected with the reduction of  $t_{sc}$  [8].

This paper deals with the study of characteristics of some scintillating plates made from composite materials and the possibility to apply heavy scintillators, including inorganic CsI : Br crystals, to improve the EM-calorimeters energy resolution.

## 2. Samples fabrication of heavy composite materials

#### 2.1. Layered composite scintillators

Process of fabrication of layered composite materials [11] is relatively simple. So, according to one of them, for the fabrication of our layered composite materials we used disks of scintillators, based on bulk-polymerized polystyrene (PS), which have the sizes of about  $\oslash$  30x4 mm<sup>2</sup>, and contain 1.5 % pTP (paraterphenyl with the maximum emission  $\lambda_{em} = 340 \text{ nm}) + 0.02 \%$  POPOP (1,4-bis-[2-(5-phenyloxazolyl)]-benzene with  $\lambda_{em} = 420 \text{ nm}$  and decay time  $\tau = 3.6 \text{ ns}$ ). On one side of the disk surfaces thin layers of a mixture of transparent epoxy glue and small inorganic scintillating crystals  $CeF_3$ ,  $Gd_2SiO_5 : Ce(GSO)^1$ , and  $Bi_4Ge_3O_{12}(BGO)^1$ , with their total thickness of h=1 mm, are deposited. Note that the absorptivity capability of these layers for  $\gamma$ -quanta is equivalent to  $h \cong 0.5$  mm layers of appropriate monocrystals. These composite materials possess an anisotropy of their optical and scintillation properties. The luminescence originating in  $CeF_3$  crystals, with  $\lambda_{em} = 340$  nm, and passing through the disks of the PSscintillators is effectively reemitted by the POPOP additive, whilst most of the luminescence originating in GSO crystals, with  $\lambda_{em} = 440$  nm, and BGO crystals, with  $\lambda_{em} = 480$  nm, remains unreemitted in our composite materials.

#### 2.2. Quasi-homogeneous composite scintillators

Experiment shows that all the attempts to introduce heavy inorganic scintillating powders of  $BaF_2$ ,  $CeF_3$ , CsI, BGO, etc. in PS-matrix resulted in the occurrence of composite materials with the light attenuation lengths  $l_o \leq 1-4$  mm, for which it is very difficult to find application in particle detectors. Our study of reasons for such low transparency occurrence in these materials showed that for obtaining an optimal level of transparency in composite materials based on plastic scintillators and inorganic scintillating crystals, it is necessary to fulfil some conditions which should be mentioned here:

— as has been established [12], in order to reach a necessary level of optical transparency in composite materials, the indices of refraction of plastic matrix  $n_p$  and introduced crystals  $n_c$  should be as much close to each other as possible; that is, an optical contrast in composite materials should be  $n_p/n_c \cong 1$ ;

<sup>&</sup>lt;sup>1</sup>These composite materials were fabricated in the Institute of Single Crystals, Khar'kov, the Ukraine

— the achievement of good mechanical and optical properties requires the provision of good adhesion, or even the creation of chemical bonds, at the boundary between dissimilar components in composite materials;

— in order to prevent unnecessary losses of transparency in composite materials, small crystals must not have fine fractions; that is, the size of introduced crystals should be as large as possible, when the natural limitation to the maximum crystal size is the thickness of scintillating plates, the homogeneity of light output, the convenience of their manufacturing procedure, etc.;

— the best shape for the introduced crystals to achieve the maximum level of transparency and density in composite materials is a small crystal ball;

— in order to secure the light output uniformity from composite materials, the light outputs of their components should be close to each other;

— in order to keep the radiation resistance of composite materials at the maximum level, the radiation resistance of introduced crystals should not be lower than the radiation resistance of its plastic matrix;

— and some others.

Our experience suggests that in order to get composite materials with the light attenuation lengths  $l_o \geq 1$  cm, first of all, the index of difference of the indices of refraction of components in a composite material should be  $\Delta n_c = |n_p - n_c| \leq 0.01$ -0.02. So, according to our measurements some  $CeF_3$  crystals and their solid solutions [13] satisfy most closely the above conditions for the creation of transparent composite materials based on PS-matrix, for example, having  $n_p \approx$ 1.5975 in the yellow wavelength region.

The main difficulty of manufacturing composite materials with good transparent properties are high values of indices of refraction for pure  $CeF_3$  crystals. They are greatly dependent on such crystals manufacturing procedures and their values are in the interval  $n_c=1.63-1.68$ , reaching the highest value  $n_c=1.68$  for very pure  $CeF_3$  crystals. Although, our measurements have shown that the index of refraction for some  $CeF_3$  crystals grown from low grade powders is about  $n_c \cong 1.6145$ . Obviously, such  $CeF_3$  crystals can provide  $\Delta n_c \cong 0.017$  for composite materials based on the PS-matrix. As for scintillation properties of such  $CeF_3$  crystals, they have the decay time  $\tau \cong 34$  ns and the light output of about  $I_o \cong 16$  % to that of a crystal anthracene. Note that these characteristics are slightly different from their typical values for  $CeF_3$  crystals,  $\tau=28$  ns and  $I_o=18\%$ .

Unlike the approach of work [12], where the index of refraction for the polymethylmethacrylate (PMMA) matrix was changed to the index of refraction of  $BaF_2$  crystals, in our case we decided to change the indices of refraction in our crystals to the index of refraction of the PSmatrix, with the help of transition from heavy crystals based on ordinary, single-component fluorides to their multi-component solid solutions with alkali and alkaline earth fluorides. This is possible because some alkali and alkaline earth fluorides [13] have the indices of refraction appreciably lower in comparison with heavy  $CeF_3$  crystals. However, for the creation of composite materials intended for the use in EM-calorimeters, not all alkali and alkaline earth fluorides are applicable. The best suited for our purposes are heterovalence solid solutions  $Ce_{1-x}Me_xF_{3-x}$ , where Me is an element from the group of the alkaline earth metals: Ba, Sr, or Ca and x is its molar fraction. Note that  $BaF_2$  (having  $n_b \cong 1.49$ ) and  $CaF_2$  (having  $n_{ca} \cong 1.44$ ) can be introduced in to these solid solutions based on  $CeF_3$  to relatively high levels of their concentrations [13], the light outputs of  $BaF_2$  and  $CaF_2$  additives are higher in comparison with  $CeF_3$  [14], and the radiation resistance of such solid solutions is over 100 kGy [15]. For the creation of composite materials we have chosen solid solutions  $Ce_{1-x}Ba_xF_{3-x}$  with  $x \leq 0.2$ . The main difficulty for the above application of these crystals is the fact that many of their vital characteristics are not thoroughly investigated. As an approximation, we suggest that the index of refraction  $n_{cb}$ , light output  $I_o$ , and averaged decay time  $\tau$  for these double solid solutions may be determined by the following equations:

$$n_{cb} \cong (1-x)n_c + xn_b,\tag{3}$$

$$I_o \cong I_{oc},\tag{4}$$

$$\tau \cong \tau_c,\tag{5}$$

where the subscripts c and b refer to the appropriate components in the double solid solutions, i.e.  $CeF_3$  and  $BaF_2$ , respectively. Taking into account the above remarks about the indices of refraction of  $CeF_3$  crystals, we estimate that the index of refraction for  $Ce_{1-x}Ba_xF_{3-x}$  will reach the required level  $n_p \cong 1.5975$ ; that is,  $\Delta n_{cb} < 0.01$ , for a  $BaF_2$  consentration somewhere in the interval 0.1 < x < 0.2. Obviously, the light attenuation length  $l_o$  in such composite materials can be  $\Delta n_c / \Delta n_{cb}$  times higher than that in composite materials based on  $CeF_3$ . Note that there is a slight variation of  $n_p$  in the PS-matrix from point to point; and  $Ce_{1-x}Ba_xF_{3-x}$  crystals are anisotropic media, i.e. their physical properties, such as index of refraction, vary with direction of light propagation. These facts are natural limitations for the transparency levels of composite materials containing these components. The experimental values of indices of refraction for  $Ce_{1-x}Ba_xF_{3-x}$  crystals at  $\lambda$ =589 nm, presented in Fig.1, show that  $n_{cb} \cong n_p$  appears to be near  $BaF_2$  consentration  $x \cong 0.115$ , and the experimental values of indices of refraction for these solid solutions begin to deviate sharply from equation (3) for  $x \ge 0.1$ .



Indices of refraction for anisotropic Fig. 1.  $Ce_{1-x}Ba_xF_{3-x}$  monocrystals as functions of  $BaF_2$  concentration x, at  $\lambda$ =589 nm.  $\blacktriangle$  — index of refraction for ordinary rays,  $n_o$  in the crystals. • — index of refraction for extraordinary rays,  $n_e$ , in the crystals. Dashed curves through our experimental points were drawn to guide the eye. A solid line shows part of n calculation according to formula (3) with the use of our experimental data, where for  $x=0 n_{cb} = n_c = 1/2(n_o + n_e) =$ 1.6145 (pure  $CeF_3$ ) and for x=1  $n_{cb} = n_b = 1.4744$  (pure  $BaF_2$ ). Horizontal dotted lines show the interval of  $n_p$  variation in PSmatrix.

Clearly, there is a keen interest to the investigation of scintillation (and optical) characteristics of such nonstiochiometric solid solutions for the above application. Figures 2b-2c depict the decay curves for some solid solutions<sup>2</sup>, where N is the counts per channel. Noticeable parts of their light outputs, ~40-45 %, have short decay times,  $\tau=11$  ns and 19 ns, respectively. The decay curve for our  $CeF_3$  crystal is presented in Fig.2a for comparison. Note that the light outputs and decay time components of these solid solutions are close to the same characteristics of the pure  $CeF_3$  crystal, which are in a good agreement with the results of [15-17], and confirm the above suggestions, expressed by equations (4-5).



Fig. 2. Decay curves for some monocrystals: (a) -  $CeF_3$ ; (b) -  $Ce_{0.95}Ba_{0.05}F_{2.95}$ ; (c) -  $Ce_{0.87}Ba_{0.13}F_{2.87}$ . The scintillators were excited by conversion electrons from <sup>207</sup>Bi.

For the creation of composite materials, the monomer was deinhibited through a column and distilled in vacuum. Then,  $100 \text{ cm}^3$  of the monomer was placed in cylindrical glass ampoules and doped with a set of scintillation additives 2 % pTP + 0.05 % POPOP. Small crystals 0-0.8 mm in size were also added to the monomer. The samples were degassed for 10-15 min and sealed off. The process of polymerization was carried out in a polysilicone bath at a temperature of about 180 °C during 24 hours.

<sup>&</sup>lt;sup>2</sup>These solid solutions were manufactured in the Institute of Crystallography, Moscow, Russia

# 3. Characteristics of new heavy scintillators

#### 3.1. Light output measurements

The 5 mm disks with diameter of about 30 mm were cut from blocks of composite scintillators. The scintillation in the samples was excited by conversion monoenergetic electrons from a  $^{207}$ Bi radioactive source, with electrons energy of about 976 keV and with an activity of about  $10^4$  Bq. The source and samples with optical contact were placed on the entrance window of a photomultiplier (PM) FEU-84-3, which had a multialkaline photocathode, with an average quantum efficiency Y = 19 % in the interval of wavelengths  $\lambda = 410-490$  nm. The photocurrent from the samples was measured in comparison with a crystal anthracene, having its thickness of about 5 mm and  $\lambda_{em} = 447$  nm. The light output of the crystal anthracene was set equal to 100 %. The experimental accuracy was determined on the base of repeated measurements, and the overall error was about  $\pm 6$  %.

Note that the light output of composite materials depends on the type of radioactive sources used for their excitation. This fact becomes especially noticeable for those of them having rather low values of the emitted particles energy. Only various kinds of minimum ionizing particles (MIP's), or equaled to them high energy particles from radioactive sources, incoming perpendicular to the sample surfaces, can give comparable, among themselves and the crystal anthracene, results of the light output definitions.

#### 3.2. Light attenuation length measurements

The measurements of light attenuation lengths  $l_o$  were carried out in accordance with the following technique. The scintillators were coupled to a PM FEU-84-3. The PM photocurrent J was measured. The scintillators were excited with a  ${}^{90}\text{Sr} + {}^{90}\text{Y}$  radioactive source, having an activity of about  $10^7$  Bq. The collimator could move along the scintillators and stop in a required position l. The measurements results were fitted by the function

$$J(l) = J_o exp(-l/l_o), \tag{6}$$

where  $J_o$  is the initial light intensity, J is the amount of light that reach the PM, and l is the thickness of the absorbing layer. The light attenuation lengths  $l_o$  were determined in the interval 0.5 < l < 2.5 cm. The accuracy of  $l_o$  measurements was about  $\pm 10$  %.

#### 3.3. Decay time measurements

The characteristic scintillation curves were measured by the single-photoelectron delayedcoincidence counting technique [18]. For these measurements PM's FEU-143 with semitransparent SbCs photocathodes and their first dynodes with  $A^3B^5$  (gallium phosphide) covering were used. The samples were excited with the help of a <sup>207</sup>Bi radioactive source. The time resolution of our experimental setup was about 0.5 ns. The measurements accuracy of decay times  $\tau_i$  and their relative intensities  $A_i$  were about  $\pm 7$  %. Note that our experimental setup did not allow us to measure decay times with  $\tau > 2\mu$ s. The experimental decay curves were fitted, after the subtraction of an accidental rate, to two (for layered composite materials containing *BGO* crystals - to three) exponentials.

Note that for the use of other radioactive sources with lower energy of emitted particles, the layered composite samples show different values for their relative intensities  $A_i$ , depending on the site of the samples excitation.

#### 3.4. Light reemission efficiencies

One of the important characteristics for new materials application in sampling EMcalorimeters [5,6] is the light intensity  $A_c$  which is reemitted from heavy scintillating plates into the 30-40 cm long WLS fibers. For this characteristic measurements we used cheap, heavy crystals  $CsI : Br [19]^3$ , with sizes 50x50x3 mm<sup>3</sup>. The crystal plates were wrapped up with aluminized Maylar. In these scintillating plates the holes 1 mm in diameter were drawn with a pitch of about 8 mm, and sixteen Y-7 [20] WLS fibers, with  $\lambda_{em} = 490$  nm, were inserted through them. The crystal plates were excited by MIP's. The  $A_c$  values were measured with the help of the PM FEU-84-3. These  $A_c$  values were compared with the same characteristic,  $A_p$ , measured for the PS-scintillating plates of the same thickness. The accuracy of these measurements was about  $\pm 8$  %.

#### **3.5.** Samples irradiation

The radiation resistance of our CsI: Br scintillators was investigated in a flux of  $\gamma$ -quanta from radioactive sources of <sup>137</sup>Cs with a dose rate of about 52 mGy/s in air at room temperature. The accuracy of the dose determination in these measurements did not exceed  $\pm 10$  %. We studied the samples transmission properties T just after the termination of irradiation (20-30 min) as well as in  $t_{rec}=1$  day of recovery in air at room temperature. The accuracy of Tmeasurements was about  $\pm 1$  %.

#### 4. Experimental results

#### 4.1. Composite materials

It is very convenient to characterize these new materials absorption efficiencies for  $\gamma$ -quanta with the help of the linear absorption coefficient  $\mu$ . Fig.3 presents the experimental and calculated dependencies of  $\mu$  for some materials as functions of the energy of  $\gamma$ -quanta in the range 10 keV<  $E_{\gamma}$  < 200 keV. For comparison, similar characteristics for a heavy polyvinyltoluen BC-452 scintillator [21] and ordinary PS-scintillator [8] are also shown in Fig.3. The calculated curves  $\mu$  for the heavy PS-scintillator [8] and new quasi-homogeneous composite material were calculated on the basis of a formula from [22]

$$\mu = \rho \sum_{i} \mu_{m,i} \omega_i, \tag{7}$$

where  $\rho$  is the medium density,  $\mu_{m,i}$  is the mass attenuation coefficient of *i* component having the mass fraction  $\omega_i$ . These experimental results show that new heavy composite materials more effectively absorb  $\gamma$ -quanta than known heavy plastic scintillators [8,20]. Since our composite materials are mechanical mixtures of their components, most of their physical characteristics are easy to calculate. So, the effective values of the density  $\rho$  and radiation length  $X_o$  for a composite material can be deduced from their known appropriate values for their components *i* 

$$\rho = \sum_{i} \rho_{i} h_{i} / \sum_{i} h_{i}, \tag{8}$$

$$X_o = \sum_i h_i / \sum_i h_i / X_{oi},\tag{9}$$

where  $h_i$  is the effective thickness of the component *i* used in the composite material.

<sup>&</sup>lt;sup>3</sup>These crystals were fabricated in the Institute of Single Crystals, Khar'kov, the Ukraine



Fig. 3. Linear attenuation coefficient  $\mu$ for some scintillators as functions of  $\gamma$ -quanta energy.  $\blacksquare$  for ordinary PS-scintillator;  $\circ$  for PS-scintillator containing 15% TPL;  $\checkmark$  — for heavy scintillator BC-452; the dashed line — calculational results for the quasihomogeneous composite material containing  $CeF_3$  crystals. Solid lines are the results of calculations with the use of formula (7).

It is well known that in the process of EM-showers development in calorimeters, there appear both the high and low energy  $\gamma$ -quanta and electrons. This fact complicates the selection of appropriate radioactive sources for the excitation of anisotropic media with the aim of accurate determination of their light outputs  $I_o$ , relative intensities  $A_i$  for  $\tau_i$ , etc. Note that the main contribution to EM-calorimeters light outputs yield high energy electrons, i.e. MIP's. In this case, the light output for an EM-calorimeter based on an anisotropic active medium under the excitation of MIP's can be easily estimated using the known values of  $I_{oi}$  and the ionization losses in components used in composite materials.

The ionization losses  $\Delta E_i$  in *i* component used in a composite material can be approximately evaluated in the following way [23]:

$$\Delta E_i \cong \alpha_i \omega_i h_i Z_i,\tag{10}$$

where  $\alpha_i$  is a constant,  $\omega_i$  and  $Z_i$  are the concentration and effective charge of atoms in the component *i*, respectively. Thus, the scintillation intensity of component *i* will be proportional to  $I_{oi}\Delta E_i$ .

Let us simplify the problem and consider that each component i of a composite material is characterized by a single (the main) set of  $\tau$  and A. In this case, the light output  $I_o$  and the relative intensities  $A_i$  for  $\tau_i$  can be calculated with the help of the following equations:

$$I_o = \sum_i I_{oi} \Delta E_i / \sum_i \Delta E_i, \tag{11}$$

$$A_i = I_{oi} \Delta E_i / \sum_i I_{oi} \Delta E_i, \tag{12}$$

where  $I_{oi}$  is the intrinsic light output of component *i*.

In Table 1 we present  $I_o$ ,  $\rho$ ,  $X_o$  and A for our composite materials, which were calculated with the help of equations (8-9) and (11-12). Some characteristics for an ordinary and heavy PSscintillators [8], containing 15 % by weight of tetraphenyl lead, TPL, are given at the beginning of Table 1.

Ν	Scintillator	$I_o$ ,	$\lambda_{em},$	$\rho$ ,	$X_o,$	$l_o$	$ au_1,$	$A_1,$	$ au_2$	$A_2$
		%	nm	$g/cm^3$	$\mathbf{cm}$	$\mathrm{cm}$	ns	%	ns	%
1	Ordinary PS-scint.	45	420	1.0	43.	40.	3.6	95	30	5
2	PSM-115+15% PTL	21	420	1.1	27.	14.	2.5	95	30	5
3	Layered PS-scint.+ $CeF_3$	22	420	1.5	12.4	3.	3.6	42	28	58
4	Layered PS-scint.+ $GSO$	31	420,440	1.6	10.8	3.	3.6	21	31	79
5	Layered PS-scint.+ $BGO$	30	420,480	1.6	8.9	3.	3.6	20	300	80
6	Comp. PMMA-scint.+ $BaF_2^*$	39	420	2.3	4.1	1.	3.6	25	650	75
7	Comp. PS-scint.+ $CeF_3$	18	420	4.3	2.4	2.	3.6	16	28	84
8	Comp. PS-scint.+ $Ce_{0.95}Ba_{0.05}F_{2.95}$	14	$420,\!435$	4.1	2.8	2.	24.	68	45	32
9	Comp. PS-scint.+ $Ce_{0.91}Ba_{0.09}F_{2.91}$	14	$420,\!435$	4.0	2.8	2.	24.	75	51	25
10	Comp. PS-scint.+ $Ce_{0.87}Ba_{0.13}F_{2.87}$	12	$420,\!435$	4.0	2.9	1.	20.	59	45	41

Table 1. Optical and scintillation characteristics of new heavy scintillators.

\* These samples of a PMMA scintillator [12] with  $BaF_2$  powder were fabricated at the Institute of Single Crystals, Khar'kov, the Ukraine.

These scintillation characteristics of our ordinary PS-scintillator are also very close to the analogous ones from [24]. As is shown in Table 1,  $X_o$  values for all the composite materials are better than those required from the active medium [8] of sampling EM-calorimeters [4,5] in order to provide them with a high energy resolution  $\sigma/E \cong 4 - 5\%/\sqrt{E}$ .



Fig. 4. Decay curves for the layered composite materials: (a) PS-scintillator plate +  $CeF_3$  crystal layer; (b) PS-scintillator plate + GSO crystal layer; (c) PS-scintillator plate + BGO crystal layer. The scintillators were excited by conversion electrons from <sup>207</sup>Bi.

The results of the decay curve measurements for the layered composite materials 3-5 are illustrated in Figs.4a-4c. The samples scintillation was excited from their sides consisting of the small crystal layers. Because of that Figs.4a-4b show all the known decay times of the components used in the composite materials. So, Fig.4a shows that sample 3 with the layer of small  $CeF_3$  crystals has  $\tau_1 \cong 3.6$  ns with  $A_1 \cong 40\%$  and  $\tau_2 \cong 34$  ns with  $A_2 \cong 60\%$ . It is well known that GSO crystals have slow scintillation rise times, and their  $I_o$  and  $\tau$  values are greatly influenced by the concentration of  $Ce_2O_3$  additive [25]. Note that in our sample 4 the used GSO crystals have  $I_o \cong 29\%$  and  $\tau \cong 130$  ns. These characteristics of our crystals differ from the reference data for GSO monocrystals, commonly having  $\tau \cong 31$  ns and  $I_o \cong 35\%$ . Due to this fact, the intensity of the second component, with  $\tau_2$ , from sample 4 containing the layer of small GSO crystals, is about  $A_2 \cong 92\%$ . The experimental results presented in Fig.4c show that for sample 5 containing the layer of small BGO crystals, the intensity of its second component, with  $\tau_2 = 60$  ns, has increased to  $A_2 \cong 27\%$ , but the intensity of its main component, with  $\tau_3 = 300$  s, has decreased to  $A_3 \cong 70\%$  in comparison with the reference data for BGO monocrystals.

These experimental results indicate that both the luminescent and scintillation properties for some small crystals (or their powders) may differ from the reference values for the same monocrystals. It is clear that under low energy type of excitations, the heavier are the crystals used in layered composite materials, the less noticeable is the intensity of the PS-scintillator component, with  $\tau_1 \cong 3.6$  ns, in comparison with their calculated values from Table 1.



Fig. 5. Pulse height spectra for the quasi-homogeneous composite materials excited by  $\gamma$ -quanta from  $^{137}$ Cs. (a) — PMMA-scintillator with  $BaF_2$  powder having  $\otimes$  11x12 mm<sup>3</sup> in size. Amplification in the pulse height analyzing channel for this material was 10. (b) — PS-scintillator with  $CeF_3$  crystals having  $\otimes 15x12$  mm<sup>3</sup> in size. Amplification in the pulse height analyzing channel for this material was 20. Arrows show approximate positions of their total absorption peaks. Dashed lines depict backgrounds.

Figures 5a-5b show the pulse height spectra from the quasi-homogeneous samples 6 and 7, excited by  $\gamma$ -quanta from a radioactive source <sup>137</sup>Cs with an activity of about 10<sup>4</sup> Bq. As is shown in Figs.5a-5b, the total absorption peak widths for these materials are enlarged due to different levels of the light outputs from the components of these composite materials.

Some characteristics of the quasi-homogeneous composite materials are summarized in Table 1. Thus, the experimental value  $I_o \cong 18\%$  for sample 7 is very close to its calculated value  $I_o = 18\%$  with the use of equation (11). The experimental results of the decay curve measurements for some quasi-homogeneous composite materials are given in Figs.6-7. Thus, Fig.7a shows that sample 7, with  $CeF_3$  crystals, has components  $\tau_1 \cong 3.6$  ns with  $A_1 \cong 21\%$  and  $\tau_2 \cong 34$  ns with  $A_2 \cong 79\%$ . Notice that these experimental results are very close to the calculated ones with the use of equation (12). It is well known that our solid solutions  $Ce_{1-x}Ba_xF_{3-x}$  also have the light emission with  $\lambda_{em} = 435$  nm [16], and the bulk of this long-wavelength emission is not reemitted by the POPOP additive of PS-scintillators. Due to high concentration levels of these solid solutions and their higher densities, the decay times of quasi-homogeneous composite materials presented in Figs.7a-7c are very close to the solid solutions given in Figs.2a-2c.



Fig. 6. Decay curves for the quasi-homogeneous composite material based on PMMA-scintillator with  $BaF_2$  powder.

The obtained values of  $\rho$  and  $X_o$  for quasi-homogeneous composite materials, samples 7-10 in Table 1, are good enough in order not to use the absorbing layers of heavy metal plates: Pb or W in EM-calorimeters [5,6]. As a result, there is a possibility to create quasi-homogeneous, i.e. with a sampling fraction close to  $f_s \cong 1$ , EM-calorimeters with a fiber-optic readout system consisting only of plates fabricated from such heavy quasi-homogeneous composite materials.

It is well known that the light attenuation in all PS-scintillators is very strong in the range of thicknesses 0 < l < 1 cm. Experiment shows that the light attenuation lengths in this region for ordinary PS-scintillating plates are  $l_o \cong 1.2$ -1.8 cm. However, for thickness l > 2.5 cm, the light attenuation lengths begin increasing to  $l_o \ge 20$  cm. Therefore, these low levels of  $l_o$  for some composite materials presented in Table 1 will not be an obstacle for their application in sampling EM-calorimeters. Moreover, materials with low values of  $l_o$  are to be used for obtaining additional information about the entrance point of incoming particles to EM-calorimeter modules. However, in this case the WLS fibers should be spaced nearer to each other than usual with a pitch of about 8-9 mm [6].



Fig. 7. Decay curves for our quasi-homogeneous composite materials: (a) PS-scintillator with  $CeF_3$  crystals; (b) PS-scintillator with  $Ce_{0.95}Ba_{0.05}F_{2.95}$  crystals; (c) PS-scintillator with  $Ce_{0.87}Ba_{0.13}F_{2.87}$  crystals. The scintillators were excited by conversion electrons from <sup>207</sup>Bi.

# 5. Heavy inorganic scintillators

Scintillating plates for sampling EM-calorimeters can be fabricated both from heavy organic and inorganic materials like: monocrystals, solid solutions, transparent glasses, ceramics, etc., provided they are cheap and posses the required optical, scintillation and radiation properties. For this investigation we have chosen cheap CsI : Br crystals [19], in order to estimate a possibility to use this heavy inorganic material for EM-calorimetry. Note that the radiation and scintillation properties of this crystal can vary. CsI : Br crystals with an optimal (from the light output point of view) concentration of CsBr exhibit better radiation and recovery properties against pure CsI crystals. Some characteristics of CsI : Br crystals are tabulated in Table 2.

	Ν	Scintillator	$I_o$ ,	$\lambda_{em},$	$\rho$ ,	$X_o,$	$l_o$	$ au_1,$	$A_1,$	$ au_2$	$A_2$
			%	nm	$\rm g/cm^3$	$\mathbf{cm}$	$\mathbf{cm}$	ns	%	ns	%
	1	CsI	18	315	4.53	1.85	1.80	10.	100		
	2	CsI:Br	45	$305,\!470$	4.50	1.84	1.80	11.	10	2000.	90
	3	$BaF_2$	46	220,310	4.89	2.05	1.49	0.8	20	650.	80
	4	ZnS:Ag	230	450	4.09	2.12	2.36	200.	100		
	5	ZnO:Ga	52	385	5.61	2.05	2.02	0.7	100		
	6	$CeF_3$	18	300,340	6.16	1.68	1.63	3.	4	28.	96
	7	GSO	35	440	6.71	1.39	1.85	31.	100		
	8	BGO	28	480	7.13	1.12	2.15	60.	9	300.	91

<u>Table 2.</u> Some characteristics of promising heavy inorganic materials that can be used for fabrication of ceramic plates.

At increased levels of CsBr concentration above its optimal level of about 2% by weight in the crystals, both the relative intensity  $A_1$  and its  $\tau_1$  begin decreasing to a weak luminescence of pure CsBr crystals with  $\tau \approx 0.07$  ns [26]. Also, fast scintillators are promising materials for EM-calorimeters supposed to work under high counting rates. Some optical and radiation properties for a CsI: Br crystal with an enhanced level of CsBr concentration of about 6% are shown in Fig.8.

Due to the hydroscopicity of the components, traces of water are often observed in the bulk of CsI : Br crystals; the water influences noticeably these crystal luminescent characteristics, i.e. in addition to the main peak of emission with  $\lambda_{em} = 305$  nm, a new light emission peak with  $\lambda_{em} \cong 470$  nm appears. A decay curve for one of such crystals, with  $I_o \cong 40\%$ , is presented in Fig.9a. As is clear from Fig.9a, this crystal has a fast component, with  $\tau_1 \cong 6$  ns and  $A_1 \cong 8\%$  and a slow component, with  $\tau_2 \cong 2\mu s$  and  $A_2 \cong$ 92%. Due to the presence of the slow component, to date these crystals have been only of limited application, i.e. only in particle detectors working under low counting rates.

A decay curve for the CsI : Br light reemitted by the Y-7 WLS fibers is given in Fig.9b. A comparison of Fig.9a and Fig.9b shows that the slow decay component with  $\tau_2$ in the reemitted light is lacking. This is possible because this slow component peak of emission does not appear to be reemitted by the



Fig. 8. Transmittance T for a 3 mm thick CsI : Br crystal as a function of the light wavelength  $\lambda$ .  $\checkmark$  — before irradiation,  $\circ$  — just after a dose of 5 kGy irradiation and  $\Box$  — after one day of recovery.

Y-7 WLS fibers. It is common knowedge that the decay curve of a scintillating plate + WLS fibers system represents a superposition of the decay times of the system components. So, with application of CsI : Br scintillating plate + WLS fibers with fast decay times, for example, BCF-92 A with a green emitting light dopant G-2 [27] having  $\lambda_{em} = 492$  nm and  $\tau = 2.64$  ns, the above system becomes operable under high counting rates up to  $\sim 5 \ 10^7 \ s^{-1}$ .



Fig. 9. Decay curves: (a) — for CsI : Br scintillating plate, (b) — for CsI : Br scintillating plate + Y-7 WLS fibers system. The scintillating plates were excited by conversion electrons from <sup>207</sup>Bi.

The measured value of light reemission intensity for the CsI : Br scintillating plate + Y-7 WLS fibers system appears to be about  $A_c \cong 0.6$  photoelectrons (p.e.)/MIP. With the use of our ordinary PS-scintillating plate in the system, the light reemission intensity appears to be about  $A_p \cong 2.0$  p.e./MIP. Note that their experimental relation,  $A_c/A_p = 0.29$ , is in a good agreement for its calculated value  $A_c/A_p = 0.20$ , taking into account their values of  $I_o$ , Tables 1-2, and  $\Delta E$ , equation (10), for these plates.

Due to low intensity  $A_1 \cong 3\%$  of its fast component, with  $\tau_1 \cong 6$  ns, this type of scintillator is usable in sampling EM-calorimeters [6,7] with the required high level of the energy resolution only for the constructions characterized by  $f_s \ge 0.3$ . To cut the EM-calorimeters cost, transparent ceramic plates can be used as an active medium. The transparent ceramic plates, being cheap, look most attractive for the above application. Note that transparent ceramics can be manufactured for a very wide class of transparent bases: oxides, fluorides, etc.

It is well known that scintillating characteristics of ceramics are close enough to appropriate characteristics of their monocrystals, if any. Only their densities  $\rho$  can be m=0.5-0.95 lower, depending on the degree of packing, from similar values of  $\rho$  for monocrystals. Obviously that increases their  $X_o$  values to 1/m times in comparison with the data for monocrystals from Table 2. Characteristics of some promising basis, which can be used for manufacturing ceramic plates for EM-calorimeters, are tabulated in Table 2. Nevertheless, their  $X_o$  are better in comparison with  $X_o$  required from the active medium [8],  $X_o=15-20$  cm, for the creation of precise EM-calorimeters [5]. Note that ceramic plates based on  $CeF_3$  or BGO can provide the radiation resistance  $\gg 100$  kGy [27,28], and plates based on ZnO : Ga can also yield EMcalorimeters fast acting under high counting rates.

# 6. Conclusion

The above experimental and calculational results disclose that with the use of new heavy scintillating materials such as active medium, there exists a feasible opportunity to create sampling EM-calorimeters with the fiber-optic readout system on the basis of the "Bayan" [6] and "Shashlyk" [5] types of calorimeter constructions. For the sampling fraction  $f_s \geq 0.5$ -0.6 [29], these EM-calorimeters [5-7] can provide the energy resolution  $\sigma/E \cong 4 - 5\%/\sqrt{E}$ , capability to work under high counting rates up to  $10^7 \text{ s}^{-1}$ , etc.

Heavy quasi-homogeneous composite materials with high densities  $\rho \geq 4.1-4.3$  g/cm<sup>3</sup> can be used for the creation of quasi-homogeneous EM-calorimeters, i.e. with a sampling fraction  $f_s \approx$ 1, with the fiber-optic readout system.

Heavy composite materials with different decay times can be used simultaneously in a single module in order to determine starting points for EM-showers development.

Heavy composite materials with low values of their light attenuation lengths  $l_o \approx 1-2$  cm can be used to obtain additional information about the entrance points of incoming particles to EM-calorimeter modules with the fiber-optic readout system.

Transparent ceramic plates based on heavy scintillators  $CeF_3$ , CsI, ZnO: Ga, etc. are very promising materials for active media application in sampling EM-calorimeters [5-7], capable of working under high counting rates up to  $10^7 \text{ s}^{-1}$ , etc.

These cheap heavy scintillators, including both layered and quasi-homogeneous composite materials, are of appreciable interest in other fields of science and technology. So, new composite materials can be used in other particle detectors, for example, in nuclear physics and medicine, for the effective registration of  $\gamma$ -quanta, neutrons, as an active element in the computer tomography [30], etc.

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Новые тяжелые сцинтиллирующие среды для прецизионных гетерогенных EM-калориметров.

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